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**FEMTOSECOND AMPLIFIERS AND MICROLASERS IN DEEP
ULTRAVIOLET**

J GARY EDEN

UNIVERSITY OF ILLINOIS

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Final Report**

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14. ABSTRACT This program pursued new sources (and amplifiers) of coherent and incoherent radiation in the ultraviolet and visible regions of the spectrum, as well as their applications in photochemistry and medical therapeutics. Research highlights include: (1) Demonstration of lasing in the visible and near-infrared from Na and Rb, respectively, by photoassociating alkali-rare gas atomic pairs, (2) Observation of the collapse of a 3-level laser system by "tuning" the energy defect between the pumped and lasing states- this resolves a fundamental question in quantum electronics, and (3) Demonstrating a new micro/nano optical amplifier well-suited for routing optical power in a scattering medium such as human tissue.					
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“FEMTOSECOND AMPLIFIERS AND MICROLASERS IN THE DEEP ULTRAVIOLET”

Prepared For

Dr. Howard R. Schlossberg
U.S. Air Force Office of Scientific Research
One Liberty Center
875 North Randolph Street
Arlington, VA 22203-1768

Submitted By

J. Gary Eden
University of Illinois
Department of Electrical and Computer Engineering
1406 West Green Street
Urbana, IL 61801

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I. INTRODUCTION AND SUMMARY OF ACCOMPLISHMENTS

This AFOSR-supported program has pursued new sources (and amplifiers) of coherent and incoherent radiation in the ultraviolet (UV) and visible regions of the spectrum, as well as their applications in photochemistry and medical therapeutics. We believe that this research effort has proven to be quite productive as evidenced by several significant accomplishments, papers published, patents received, and the founding of two companies. The most prominent research highlights resulting from this grant are:

1. Observation of lasing in the visible and near-infrared from Na and Rb by photoassociating Na-Xe and Rb-Xe atomic pairs, respectively. The laser excitation spectra for this pumping process have been measured and, in recent experiments, an internal “yardstick” for measurements of absolute cross-sections for the photoassociation of alkali-rare gas atomic pairs, relative to those for the two photon excitation of the alkali atom, has been demonstrated.

2. The collapse of a three-level laser system has been observed directly in experiments in which the energy defect (ΔE) between the lasing and pumped excited states is “tuned” and approaches kT . This resolves a fundamental question in quantum electronics and contradicts a long-standing assumption in the laser community.

3. A new micro/nano optical amplifier has been developed. Collaboration with another research group at Illinois has yielded an amplifier that is injection-locked by an internally-generated Raman signal. This new amplifier appears to be ideally suited for routing optical power through a strongly scattering medium such as human tissue.

4. Arrays of microchannel plasmas have been shown to be efficient in generating ozone or dissociating CO_2 . The former has led to the founding of a company (EP Purification) devoted to the disinfection of water with ozone produced in reactors comprising arrays of microplasmas.

The highlights described above are only a few of the accomplishments realized under this program. With regard to other tangible results, 31 scientific articles have been published in peer-

reviewed journals. Also 23 patents, both U.S. and foreign, have been granted during this program. It must be emphasized that all of these (publications and patents) acknowledge AFOSR support and most were made possible *solely* by AFOSR. In the next section, a few selected accomplishments are described in greater detail. Before presenting these results, however, let me personally express my thanks to AFOSR and Dr. Schlossberg, specifically, for the continuing support that has allowed for our students to be educated in areas of interest to the DOD, and for developing technologies that are responsible for the founding of three companies. I am pleased to report that the companies have already created 16 full-time jobs and, for this reason as well, we are grateful to AFOSR for its support!

II. OVERVIEW OF RECENT ACCOMPLISHMENTS: ALKALI LASERS PUMPED BY COLLISION PAIR EXCITATION, AND RARE GAS DIMER DYNAMICS IN PLASMAS

This section presents results from highlights of several experiments that were mentioned briefly in the last section.

A. Dual Wavelength Sodium Laser Pumped By Na-Xe Pair Absorption

Laser guide stars have applications of growing importance to the DOD, astronomy, and environmental monitoring. However, realizing a laser that is automatically “locked” to one of the D-lines of an alkali atom has been challenging in the past and existing guide stars are, not surprisingly, quite sophisticated (owing to the nonlinear conversion steps required) and expensive. Building on earlier work in our laboratory in which we demonstrated lasing in Cs and Rb, we have recently obtained lasing simultaneously on both the D-lines of Na. Figure 1 is a low resolution spectrum

in the yellow (588.9-589.7 nm) showing lasing on the $3^2P_{1/2, 3/2} - 3^2S_{1/2}$ (D_1 , D_2) lines of Na when Na-Xe pairs are photoassociated at 560 nm. It is interesting that the 588.9 nm line exhibits higher intensity than its 589.6 nm partner, despite the fact that the $3^2P_{3/2}$ state degeneracy is

twice that for the $^2P_{3/2}$ state. Thus, the 589.6 nm laser appears only because of Na*-Xe collisions that

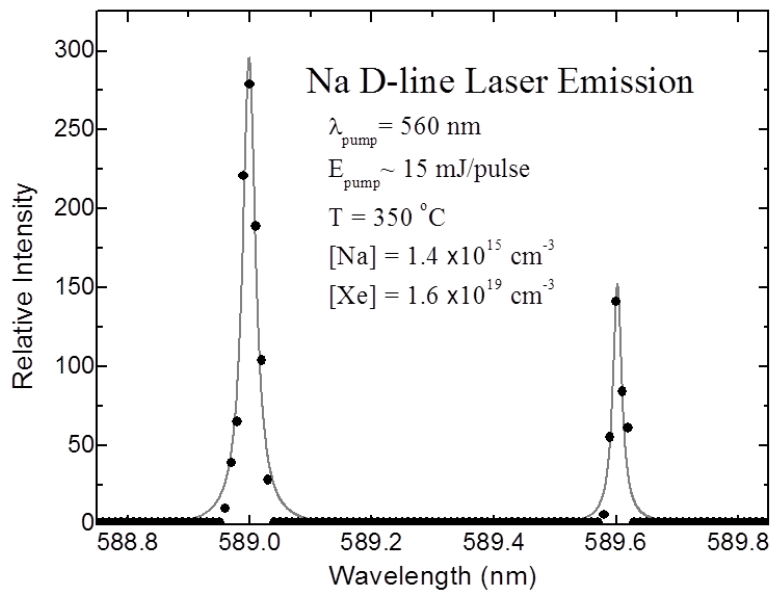


Fig. 1 Low resolution laser spectrum in the yellow, obtained by photoassociating Na-Xe atomic pairs. Simultaneous lasing on the 489.0 nm and 589.6 nm lines of Na is observed.

populate the $3^2P_{1/2}$ level. A somewhat higher resolution view of the D₂ laser spectrum is presented in Fig 2. Obtained with a scanning monochromator, this spectrum has a FWHM of $6.5 \cdot 10^{-2} \text{ nm}$ which is limited by the resolution of the detection system. The actual linewidth for

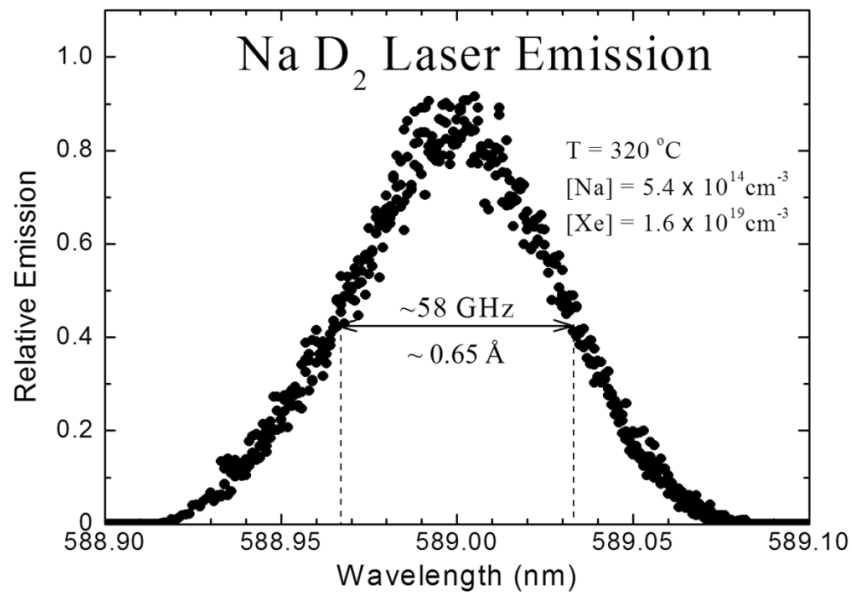


Fig. 2 D_2 (589 nm) laser spectrum obtained by photoassociating Na-Xe pairs at 560 nm. The resolution of the spectrum ($6.5 \cdot 10^{-2}$ nm) is instrument-limited.

the 588.9 nm (D_2 line) laser was measured recently with an etalon and the resulting fringe pattern is shown in Fig. 3. The measured linewidth of 8 ± 1 GHz suggests that the lasing species is not the Na atom itself but rather Na^* -Xe pairs having an internuclear separation near the peak of the nearest neighbor distribution (~ 2.2 nm for a Xe background density of $1.6 \cdot 10^{19} \text{ cm}^{-3}$).

Confirmation that the pumping process responsible for Figs. 1 and 2 is, indeed, the photoassociation of Na-Xe pairs is provided by Fig. 4 which compares the D_2 laser excitation spectrum (lower panel) with the blue satellite absorption spectrum published by Chung *et al.* [1]. Note that our excitation spectrum coincides nicely with the strongest undulation in the Na-Xe free-free absorption spectrum. Because this laser pumping scheme provides output that is “locked” to an atomic transition while allowing considerable flexibility in the pump wavelength, photoassociation-pumped lasers hold considerable promise as guide star lasers. The most exciting aspect of these results, however, is that both the laser excitation spectrum (Fig. 4) and the étalon pattern (Fig. 3) show the unmistakable influence of Na-rare gas pairs. That is, the Na^* -Xe complex is the critical species in both the pumping and lasing processes and yet its impact is detected only indirectly.

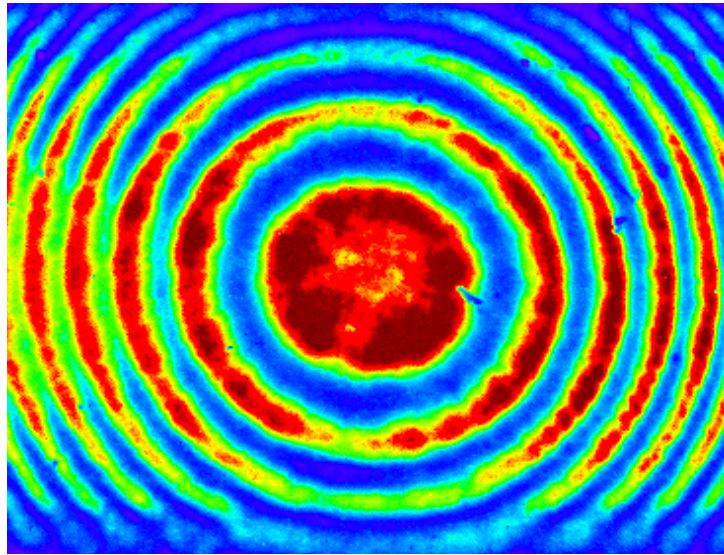


Fig. 3 Fabry-Perot étalon pattern observed for the Na 588.9 nm (D_2 line) laser. This fringe pattern yields a linewidth of 8 ± 1 GHz.

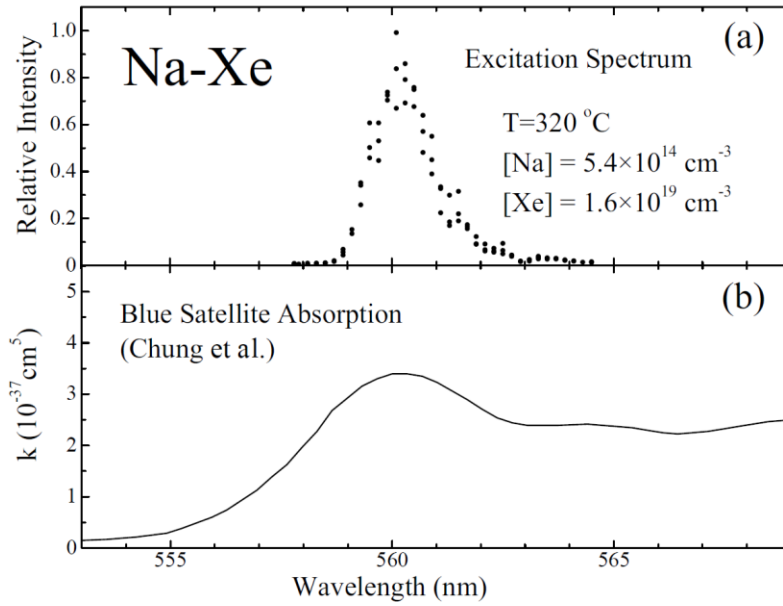


Fig. 4 Laser excitation spectrum for the Na D₂ laser (lower panel) compared with the Na-Xe absorption spectrum recorded over the same region [1].

B. Transformation Of a Three Level Laser Into a Two Level (Non-Lasing) System

A longstanding and fundamental question in laser and maser physics is that of the minimum energy separation (ΔE) between the pumped and lasing states in a three level system. We have addressed this question with the diatomic system of Fig. 5. This diatomic model allows for ΔE to be tuned continuously without altering any of the other parameters of the laser system. Photoassociating Cs-Ar atomic pairs at various wavelengths while monitoring the Cs D₂ line (852.1 nm) yields the laser excitation spectra shown in Fig. 6 for several different temperatures. Note that the low ΔE portion of the spectra of Fig. 6 is suppressed as the temperature is increased, a result due to tighter thermal coupling between states 2 and 3. If the temperature continues to be raised to 483 K (Fig. 6), the three level laser system collapses. The transformation of a 3-level laser into a two level (non-lasing) system is quantified in Fig. 7 which shows the variation with temperature of two measures of the strength of the excitation spectrum

(which is related to the population inversion ΔN). It is clear from Fig. 7 that when ΔE falls below ~ 0.7 kT, the 3-level laser ceases to function.

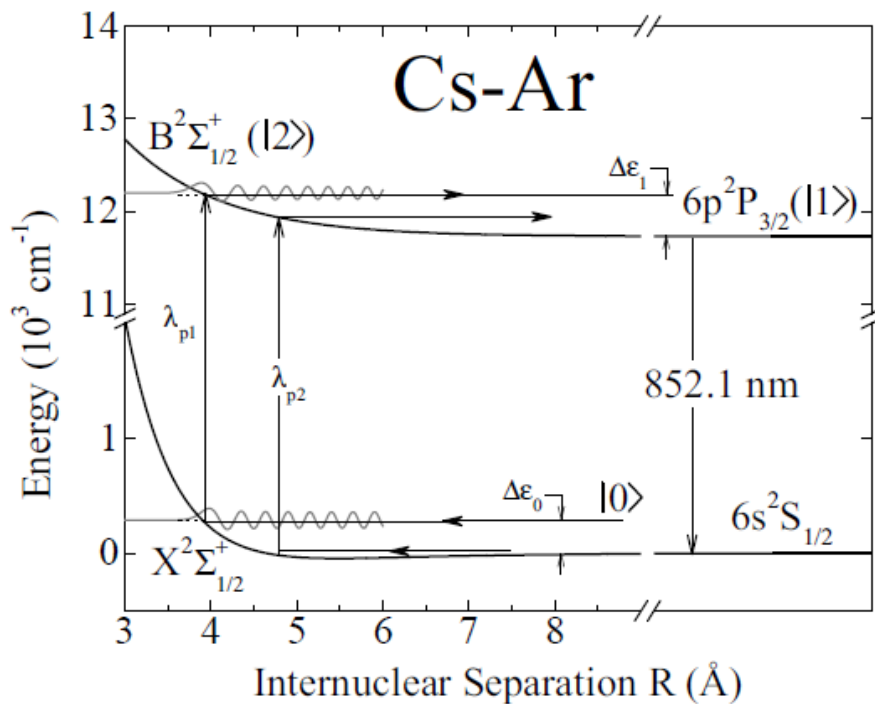


Fig. 5 Partial energy diagram for Cs-Ar pairs, illustrating the generation of lasing on the Cs D_2 line (852.1 nm) following the photoassociation of the Cs-Ar complex.

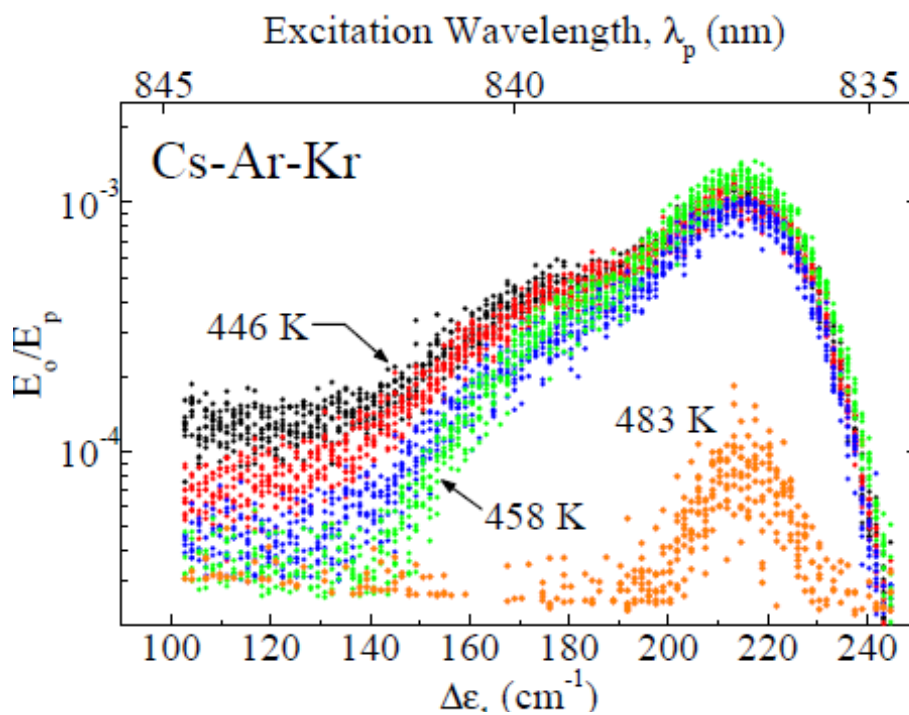


Fig. 6 Cs D₂ (852.1 nm) laser excitation spectra acquired with a Cs-rare gas mixture.

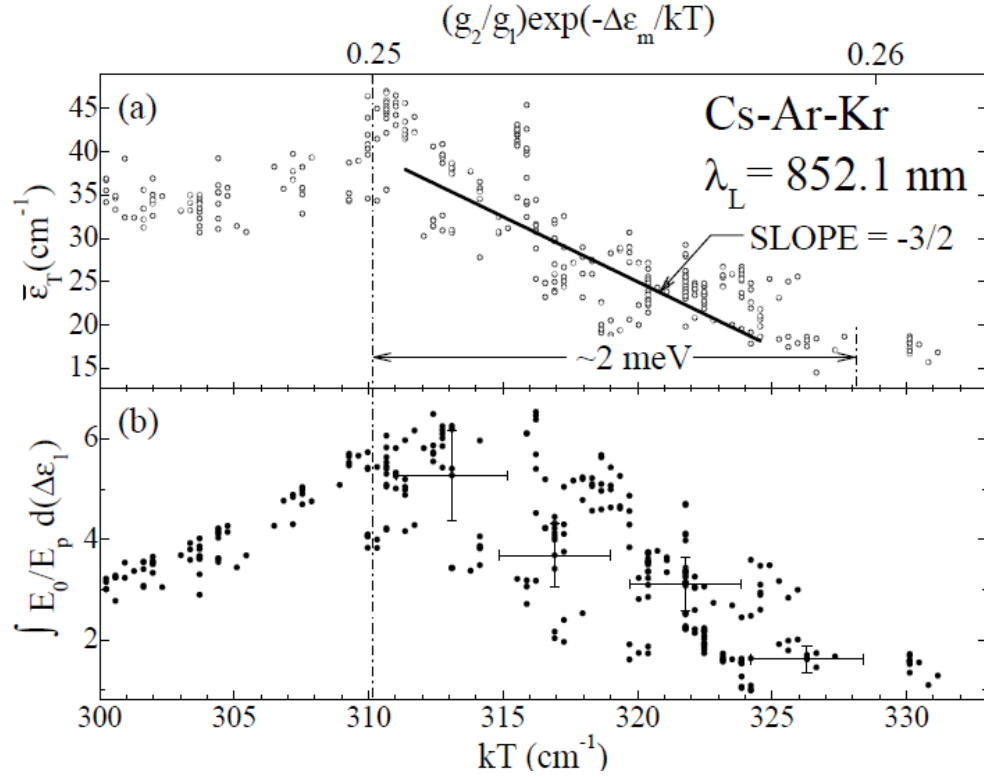


Fig. 7 Dependence on thermal energy (kT) of two measures of the laser excitation spectrum strength. The upper panel presents the exponential decay constant associated with the red portion of the spectra of Fig. 6, and the lower panel shows the wavelength-integrated values for each spectrum

We note that the Boltzmann factor for this value of ΔE is $1/2$. We believe this to be a fundamental result with implications for laser design. Note that the data for Figs. 6 and 7 were obtained by photoexciting Cs-Ar-Kr *trios* of atoms, as opposed to the alkali-rare gas pairs of previous work. The properties of the Cs*-Ar-Kr complex clearly defines the characteristics of the laser of Fig. 5.

Data similar to those of Fig. 4(b) for the Rb-Xe collision complex are shown in Fig. 8. As was the case with the Cs-Ar-Kr experiments, lasing on the D₂ line of the alkali (Rb) is monitored as the thermal energy kT is increased. For this system, the collapse of the three level laser is clearer than was the case for the Cs-rare gas system. It is also interesting that the width of the transition from stable operation of the 3-level laser to termination of lasing is narrow, ~ 2 meV.

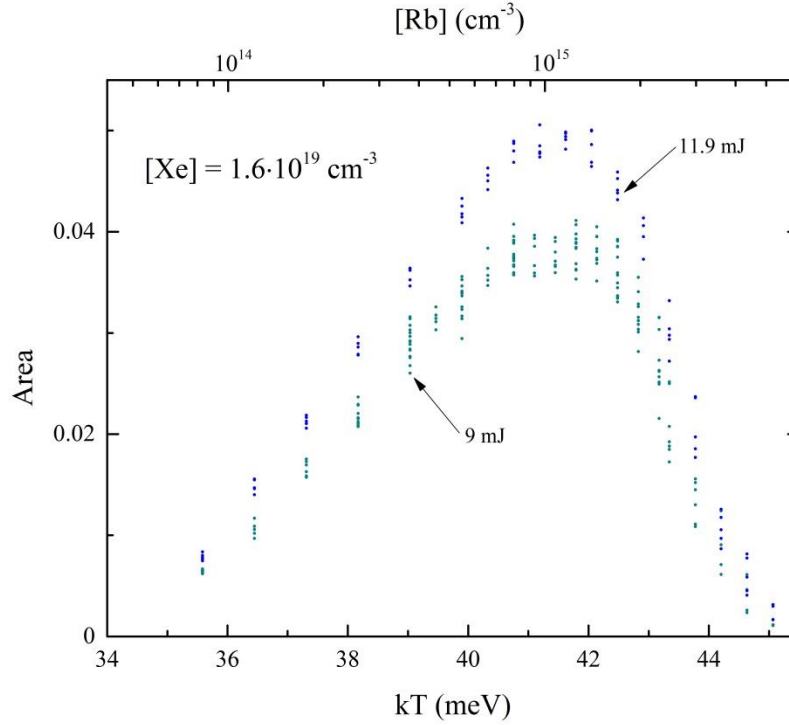


Fig. 8 Data analogous to those of Fig. 7 for the Rb-Xe system. Lasing on the D₂ line of Rb is monitored as kT is increased, and data are shown for two values of the laser pump pulse energy.

C. Cooling of Electronically-Excited Molecules in a Microplasma Jet

The ability to cool molecules to ~4 K in a supersonic jet has been known for decades. To date, however, it has been possible to cool only the ground state species but it is the excited states that are of greatest interest as emitters. At Illinois, we have recently succeeded in integrating a microcavity plasma device with a micromachined supersonic nozzle which allows us to produce specific excited molecules directly in the supersonic expansion. The result is that we have been able to demonstrate cooling of electronically-excited molecules. Figure 9 is a spectrum shown in false color, produced by a microcavity plasma that is generated in a parabolic cavity through which He gas flows (driven by a pulsed solenoid). The specific spectrum presented in Fig. 9 is that of the $e^3\Pi_g \rightarrow a^3\Sigma_u^+$ transition of He₂ in the blue (~465 nm).

Obtained with an imaging spectrometer, this spectrum shows clearly that, despite its short lifetime of ~ 200 ns, the $\text{He}_2(e)$ state molecules are cooled to $< 35\%$ of their initial kinetic

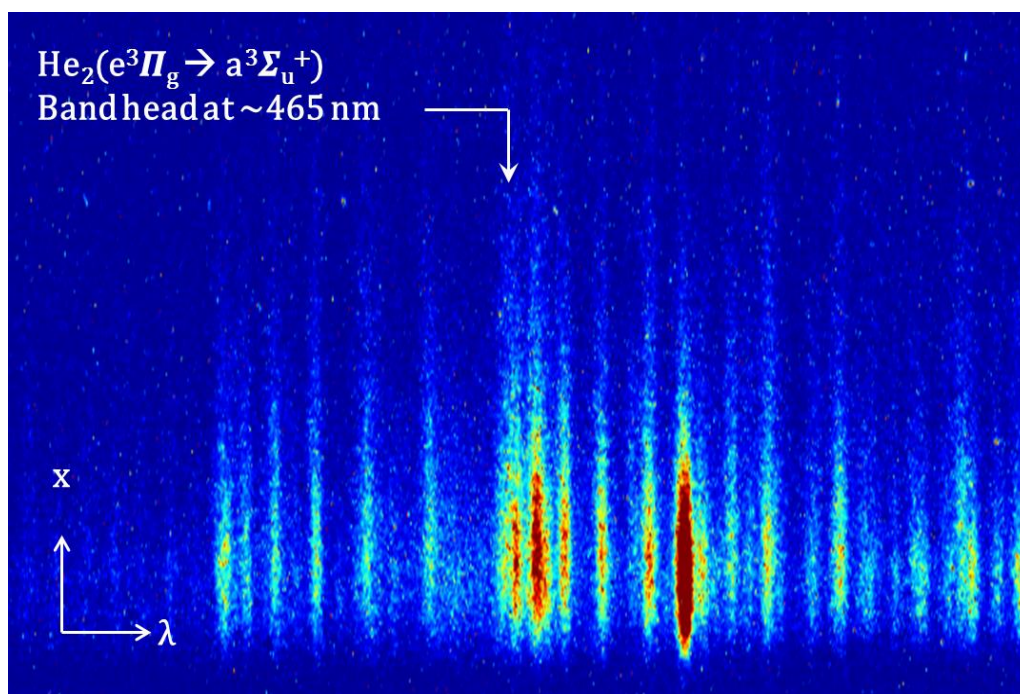


Fig. 9 Emission spectrum of $\text{He}_2(e \rightarrow a)$ emission produced in a microcavity plasma integrated with a supersonic nozzle.

energy. This result and others obtained to date show that cooling of a wide range of molecules can now be accomplished. This represents a powerful tool for molecular spectroscopy and laser development.

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